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NATURAL BIOPOLYMER BASED ELECTROLYTES FOR DSSC APPLICATION : A REVIEW

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ABSTRACT

The world's traditional energy sources, fossil fuels, are rapidly running out because of the enormous demand for energy, population growth, and the advancement of civilization. Our environment is directly impacted negatively by these energy sources. Solar energy, for example, converts light energy directly into electrical energy without causing any negative impacts on the environment, which is why its use has drawn a lot of attention. DSSC is one of the various types of solar cells that have been used to harvest solar energy. One of the primary components of its construction is electrolyte, along with other components. Because of their sustainability, large supply of raw materials, nontoxicity, low cost of production, ease of processing, and low volatilization when compared to liquid-state electrolytes, researchers have given natural biopolymer-based electrolytes a lot of attention. An overview of the most recent developments in the creation of biopolymer-based DSSCs is the goal of this review.

KEYWORDS: Solar Energy, Solar Cells, DSSC, Electrolyte, Biopolymer

INTRODUCTION

Every day, there is a greater and greater need for energy due to enhanced modern civilization along with a growing population. The world's fossil fuels—which provide almost 80% of the energy needed today—are running out rapidly **[1]**. Therefore, one of our top priorities should be to find sustainable alternative energy sources. For the last few decades, researchers have been working to investigate renewable energy sources. One of the most promising renewable energy sources is photovoltaic conversion of solar radiation.Solar cells have been studied the most, along with other power sources including hydro, wave, tidal, wind, etc. From the very beginning of its initial construction by Gratzel et al. in 1991, the dye-sensitized solar cell (DSSC) has gained significant attention as one of the most promising methods for converting solar energy into electrical energy **[2]**. The operation of DSSCs closely resembles that of plant photosynthesis.This technology of DSSCs is highly appealing because of its ease of manufacturing, low raw material costs, environmental-friendly material utilization, processing techniques using basic and affordable equipment, and the potential to create flexible solar cells. Electrolyte materials, one of the main parts of DSSCs, continuously regenerate the dye and themselves while also handling the inner charge carrier transfer between the photoanode and the counter electrode **[3]**. Redox couples and a variety of metal-salt-based liquid electrolytes and ionic liquids have been employed in DSSCs since the outset. Yet, issues with leakage, organic solvent volatilization, flammability, flexible cell design limitations, electrode photocorrosion, and liquid electrolytes' long-term electrochemical instability have shifted research attention to how to replace liquid electrolytes with solid alternatives. The usage of polymer-based electrolytes, which are less expensive, easier to prepare, sustainable, and environmentally benign, is becoming more popular in this context in an effort to increase overall efficiency and discover innovative solutions that might solve the issues mentioned above. There have been reports of the use of a wide variety of ionic liquid-based electrolytes, biopolymers, aqueous synthetic polymers, and their derivatives in the production of DSSCs **[4-6]**.

However, the majority of the synthetic polymers used in DSSCs do not degrade when exposed to microbes, which causes environmental issues and compels scientists to concentrate on biodegradable polymers. During the past 20 years, research on the use of biopolymer-based electrolytes in the DSSC has grown exponentially. Cellulose, chitosan, agarose, carrageenan, and starch are examples of natural biopolymers that have been widely employed as electrolytes in DSSCs **[7-11]**. Biopolymer electrolytes are attractive candidates for DSSCs because of their reasonable conductivity, strong thermal stability, and capacity to create biodegradable films. This review will discuss the latest developments in using natural biopolymers (polysaccharides) as electrolytes in DSSCs.

2. STRUCTURE OF DYE SENSITIZED SOLAR CELLS (DSSCS)

Transparent conductive oxide (TCO), semiconductor metal oxide (SMO) or photoanode, dye sensitizer, electrolyte, and counter electrode are the five general components of DSSCs.

2.1. Transparent conductive oxide (TCO)

In addition to supporting cell components and serving as a gateway for solar radiation absorption into solar materials, the transparent conductive oxide substrate (TCO) gathers and transfers produced electrons to the counter electrode without reducing energy loss. It should therefore have low electrical resistivity and be extremely optically clear. The most widely utilized TCO materials in DSSCs are fluorine-doped tin oxide

(FTO) glass, aluminum-doped zinc oxide (AZO), and indium-doped tin oxide (ITO) glass **[12]**.

2.2 The Photoanode

A semiconductor metal oxide (SMO) sensitized with dye molecules is mainly known as the photoanode. TiO2, ZnO, SnO2, Nb2O5, or CdSe are examples of the favored photoelectrodes. To create flexible solar cells, however, metal oxides have been replaced by plastic foil, inorganic thin films, organic inorganic films, and certain metal sheets **[13]**.

2.3. Dye Sensitizer

Covalent bonds are formed between a monolayer of dye molecules, referred to as dye sensitizer, and the semiconductor metal oxide (photoanode) of DSSCs **[13]**. The dye sensitizer interacts directly with electrolytes and semiconductor materials both electrically and physically. After absorbing solar light, it discharges photoexcited electrons to the SMO at the dye/SMO contact, which then transmits them to the TCO.

2.4. Electrolyte

By absorbing electrons from redox mediators, the electrolytes in DSSCs play a crucial role in recovering oxidized dye molecules to their neutral state. It is primarily made up of a redox pair, such as I^-/I_3^- , $SCN/(SCN)_3^-$, S/S_2^-, and Co^{2+}/Co^{3+}, which is placed between two electrodes and contains organic matrix (solvents and certain polymers to improve physical, optical, electrical and thermal properties). Higher electrical conductivity and diffusivity, long term chemical, electrochemical, optical, thermal, and interfacial stability, reduced absorption of solar radiation in the visible to near-infrared spectrum, and the absence of dye molecule desorption and degradation from the oxide surface are all considered important requirements for the perfect electrolyte composition **[3]**. Iodide/triiodide (I^-/I_3^-) redox couples are the best option for the realistic DSSC construction when all those parameters are taken into consideration. Electrolytes can be divided into three categories based on their composition, physical states, and modes of operation: (i) liquid electrolytes, (ii) quasi-solid electrolytes or gel electrolytes and (iii) solid electrolytes.

2.5. Counter Electrode (CE).

Counter electrodes (CE) are simply a metallic-coated transparent conductive layer (TCO) **[14]**. Their primary roles are as follows: (i) they act as a catalyst to provide electrons for the redox reaction to be completed, reducing oxidized dyes by collecting electrons through electrolytes (a mediator); (ii) they act as a positive electrode to gather electrons from the external circuit and return them to the circuit to finish the circulation; and (iii) they act as a mirror to reflect light that has not been absorbed by the cell and return it to the cell and enhancing cell performance.

3. CLASSIFICATIONS OF DSSC ELECTROLYTES

3.1. Liquid Electrolyte

The three primary constituents of liquid electrolytes are additives, ion conductors, and solvents **[15]**. Primarily organic in nature, the solvents facilitate the dissolution of ionic conductors and regulate the diffusion between the electrolyte and semiconductor and CE. Therefore, a solvent with a high dielectric constant, high conductivity, and low viscosity is ideal. Ionic liquids (ILs) have several benefits over organic solvents, including greater chemical and thermal stability,lower volatility, and comparatively non flammability.

3.2. Quasi-Solid State or Gel Electrolytes

Although liquid electrolytes can be used to create very effective DSSCs, they have some practical issues, such as dye photodegradation, counter electrode corrosion, and short life in addition to leakage and quick solvent volatilization. These issues can be avoided by using quasi-solid-state or gel electrolytes in DSSC fabrication. The quasi-solid state electrolyte is an attractive replacement for liquid electrolytes due to its adjustable physical characteristics, sealing, and durability **[16, 17]**. This gel electrolyte possesses the diffusivity of liquid electrolytes and the cohesiveness of solid electrolytes **[18]**.

3.3. Solid State Electrolyte

The solid-state electrolyte is regarded as another efficient substitute for the liquid electrolyte because it can attain great mechanical stability and virtually no leakage issues with simpler synthesis procedures. The solid state electrolyte based DSSC is composed of a p-type sensitizer and an n-type semiconductor material, similar to the concepts of Si-based p-n junction solar cells.

4. BIOPOLYMERS IN THE DSSCS ELECTROLYTE PREPARATION

The most popular biopolymer among the various biopolymers is polysaccharides such as carrageenan **[10, 19-22]**, cellulose **[8, 9]**, agarose **[11]**, starch **[9, 23-26]**, chitosan **[8, 24, 27-30]**, and several of their water-soluble and organo-soluble derivatives are widely utilized as electrolytes in DSSCs. **Figure 1** shows the chemical structures of the various polysaccharides and their derivatives that are utilized to create DSSC electrolytes.

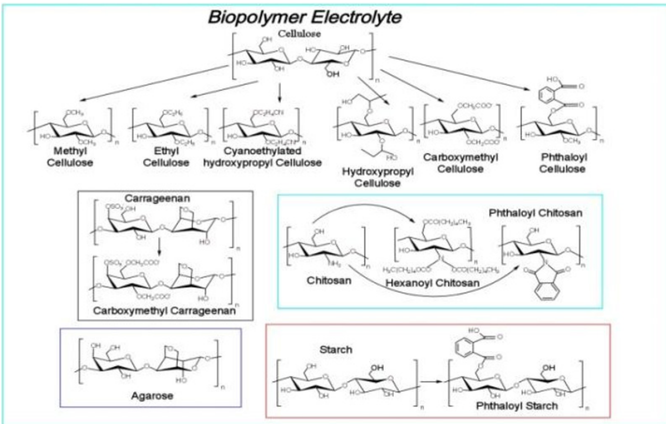

Figure 1: Chemical structures of some typical naural biopolymers and their derivatives that are utilized as electrolytes in DSSCs.

4.1 Cellulose and Its Derivative-Based Electrolytes

According to **Figure 1**, cellulose is the most prevalent naturally occurring renewable biopolymer composed of the β-(1−4)-

glycosidic linkages of D-glucose units. Since this chemical structure contains several hydroxyl groups, it produces a linear polymer with a high polar surface **[31]**. It is a great option for the production of inexpensive and environmentally friendly electronic devices like DSSCs because it is biodegradable and both chemically and thermally stable **[32]**.

4.1.1. Cellulose-Based Electrolytes

There have been reports of using cellulose that is microcrystalline, microfibrillated, nanocrystalline, and nanofibrillated as a gel electrolyte system for DSSCs .They can be made from cellulose fibers that have been removed using a variety of chemical, mechanical, or enzymatic processes **[33]**. By removing the intricate procedure required for previous cellulosic material preparations, Salvador and his team used unmodified microcrystalline cellulose (MCC) to create more affordable and environmentally friendly DSSCs **[34]**. However, Bella et al. created a cell that consists of a nanoscale microfibrillated cellulose (NMFC) biopolymer electrolyte and a cellulosic fiber-based photoanode in an effort to create paper-based DSSCs **[35]**. The effectiveness of their paper-based DSSC was 3.55% and 5.20% under 1 and 0.2 solar irradiation, respectively.

4.1.2. Cellulose Derivatives-Based Electrolytes

Highly crystalline pure cellulose was used in earlier research on cellulose electrolytes. Because of the high crystallinity of cellulose, DSSC electrolytes had low efficiency and conductivity **[36]**. Nevertheless, grafting, plasticization, mixing with another polymer, and chemical changes can all lessen this crystallinity **[37]**. In the synthesis of DSSC electrolytes, a variety of cellulose derivatives that are both water-soluble and organosoluble have been employed.

To manufacture crack-free photoanode ZnO film for DSSC, for instance, ethyl cellulose (EC), a cellulose derivative, was employed as an organic binder **[38]**. The microcracks in the ZnO film were lessened by the addition of 10 weight percent EC, increasing the fill factor and, consequently, the DSSC's efficiency to 3.8%.

Another derivative of cellulose, hydroxypropyl cellulose (HPC)-based gel electrolyte DSSC, showed good performance because of the presence of bulky functional groups. Khanmirzaei et al. reported 3.94% efficient DSSCs with an electrolyte consisting of HPC and NaI, containing ethylene carbonate and propylene carbonate as plasticizers **[39]**. Furthermore, an even more efficient DSSC of 7.55% efficiency was prepared with an acrylonitrile derivative of HPC named cyanoethylated hydroxypropyl cellulose (CNHC) **[40]**.

On the other hand, Bella and colleagues made a biopolymer gel electrolyte of PEO and CMC together with a liquid electrolyte of MPII and NaI dissolved in acetonitrile for the creation of DSSCs **[41]**.

4.2. Agarose-Based Electrolytes

As shown in **Figure 1**, agarose, also known as agar, is a linear polymer made up of alternating β-D-galactose and 1−4-linked 3,6-anhydro-α-L-galactose units that is extracted from seaweed. It is commonly employed as an aqueous electrolyte gel. An electrolyte based on agarose demonstrated conductivity between about 10^{-2} and 10^{-3} S/cm, making it a viable polymer matrix for the production of DSSCs **[42]**.

4.3. Chitosan and Its Derivatives-Based Electrolytes

A linear polyaminosaccharide, chitosan is the N-deacetylated form of chitin, the second most prevalent biopolymer **[43]**, which is composed of residues of 2-acetamido-2-deoxy-D-glucopyranose and β-(1,4)-2-amino-2-deoxy-D-glucopyranose **(Figure 1)**. At ambient temperature, pure chitosan has a conductivity of 10^{-10} to 10^{-9} S/cm **[44]**, but a swollen chitosan membrane has a conductivity of 10^{-4} S/cm **[45]**. For various electrical applications, its high conductivity in the hydrated state, bioavailability, nontoxicity and good complexity with ionic liquids make it a desirable material **[46]**. Chitosan's aforementioned qualities have drawn a lot of attention to its use in the manufacture of DSSC electrolytes **[24]**.

4.3.1. Chitosan-Based Electrolytes

A chitosan-based electrolyte gel was created by Singh et al. using NaI/I_2 redox mediator and EMISCN ionic liquid (IL) **[47]**. Effective incremental IL incorporation increased the gel's smoothness and amorphic character, which in turn improved conductivity. By adding NH_4I, 1-butyl-3-methylimidazolium iodide (BMII) ionic liquid, and ethylene carbonate (EC) plasticizer to the matrix, Buraidah et al. **[48]** further enhanced the conductivity and performance of N3 colored DSSCs built of chitosan-based electrolytes by multiple times. The effectiveness of using natural dyes instead of synthetic N3 dye for chitosan-based DSSCs was investigated by Buraidah **[7]** and Boonchaisri **[49]**. The results showed that DSSCs made of N3 dyes were more efficient than those made of natural dyes due to the chemical structure of N3 dyes and their greater capacity to inject electrons. Similarly, Yahya et al. combined chitosan with poly(vinylidene fluoride hexafluoropropylene) (PVF-HFP) using $MPII/KI/I^2$ electrolyte to create a highly conductive gel **[27]**. On the other hand, Khalili et al. prepared a gel DSSC electrolyte with chitosan nanoparticles (CSNP) and water-based chitosan using the ionic gelation and ultrasonication methods **[29]**. When CSNP was used in place of chitosan in electrolytes, the efficiency was better because the smaller size of CSNP provided a lower resistance at the counter electrode/electrolyte interface, increasing ion mobility and resulting in a higher JSC. In addition to these electrolytes, Jin et al. **[30]** have employed chitosan as a binder for a TiO_2 photoelectrode. Chitosan's incorporation improved pore distribution and decreased pore sizes, which changed the TiO_2 surface's shape. By shortening the electron transit time and lengthening the recombination period, a mere 2% chitosan addition on the TiO_2 surface raised the efficiency to 4.18%.

4.3.2. Chitosan Derivatives-Based Electrolytes

Chitosan's crystalline structure, structural rigidity and intermolecular H-bond formation caused it to be insoluble in various common organic solvents and soluble only in diluted acidic solutions **[50]**. Buraidah et al. created organo-soluble and higher ionic characteristic phthaloyl chitosan (PhCS) via the

simple substitution of phthalic anhydride for hydrogens in the amino functional groups of chitosan, and they investigated its suitability for DSSCs **[28]**. Phthaloylation of chitosan enhanced the DSSC's conductivity and efficiency due to the extra ionic character.

Semicrystalline polymers can be taken into consideration for ion conduction since they have more polymeric segmental movements than crystalline polymers, which perform the worst in this regard **[51]**. Muhammad et al. created a combination of semicrystalline hexanoyl chitosan (HC) and an amorphous PVC-based electrolyte for solid state DSSC in consideration of this characteristic **[52]**. In comparison to pure HC and PVC, blending produced a stronger, less crystalline polymer host with better conductivity when NaI was present.

4.4. Starch and Its Derivative-Based Electrolytes

Due to its mechanical stability and reduced steric hindrance for the mobile ions, starch, a carbohydrate biopolymer, has drawn a lot of interest as an electrolyte for DSSC **[53]**. In order to create DSSC electrolytes, starches with a high amylose content, such as sago, arrowroot, rice, and potato starches, have been widely utilized. Chemical structures of starch along with its derivative phthaloyl starch are shown in **Figure 1**.

4.4.1. Starch-Based Electrolytes

Khanmirzaei and his team have thoroughly investigated the potential use of RS(rice starch) with a high amylose content and low viscosity as an electrolyte for DSSC manufacturing **[25,53]**. In a study, they created a polymer electrolyte for DSSC using biodegradable rice starch combined with LiI solution. The results of this investigation demonstrated that adding LiI salt made the composite more amorphous, which raised its conductivity. By adding 35 weight percent LiI to the mixture, the conductivity of the rice starch electrolyte increased from 6.87×10^{-10} to 4.87×10^{-5} S/cm [53]. Similar to this, Yogananda et al. have reported creating DSSC using rice starch as the water-based gel electrolyte [25]. They have separated and refined rice starch nanoparticles and created an electrolyte by adding LiI and I_2 to the starch solution, which was utilized to create DSSC. The cell's photovoltaic performance was more efficient than its liquid electrolyte of LiI and I_2, with an efficiency of 0.35%. Additionally, Singh et al. discovered a biodegradable electrolyte derived from arrowroot and sago palm starch in addition to starches from rice and potatoes **[54].**

4.4.2. Starch Derivatives-Based Electrolytes

In addition to reducing viscosity and crystalline properties, several chemical modification techniques, including grafting, esterification, and oxidation, can be utilized to improve thermoplastic and thermal stability. A higher amorphous modified cation potato starch (CPS) was created, for instance, by Lobregas et al. grafting 1-glycidyl-3-methylimidazolium chloride (GMIC) on the backbone of the raw potato starch **[26]**. In order to get around the need for water as a solvent for starch-based electrolytes, phthaloyl rice starch (PhRS), an organic soluble esterified starch derivative, polymer, has recently been employed for the DSSC electrolyte production **[9,23]**. To create a gel, PhRS was mixed with hydroxyethyl cellulose (HEC), TPAI/I_2 was used as a redox mediator, and DMF(dimethyl-formamide) was used as a solvent. Along with higher mechanical strength, the produced gel demonstrated good conductivity when compared to pure starch-based electrolytes.

4.5. Carrageenan and Its Derivative-Based Electrolytes

Carrageenan is a class of linear sulfated polysaccharides that are derived from red edible seaweeds and consist of D-galactose and 3, 6-anhydro-D-galactose **(Figure 1) [10]**. Depending on how many and where the ester sulfate groups are found, there are three forms of carrageenans: (a) lambda (λ) ($C_{12}H_{19}O_{20}S_3^{-3}$), (b) kappa (κ) ($C_{24}H_{36}O_{25}S_2^{-2}$), and (c) iota (ι) ($C_{24}H_{34}O_{31}S_4^{-4}$) **[55]**.This polymer functions as an anionic polyelectrolyte due to the SO_3 − **ion group in its structure. From the standpoint of solid-state electrochemistry, it can also function as a great polymer host by offering numerous coordination sites for the transportation of metal cations [56]**. In addition to serving as a carrier for the charged salt species, oxygen atoms in the polymer can also form coordinate bonds with the cations of the redox mediators that cells use **[22]**. Furthermore, carrageenan's high hydroxyl group content facilitates the formation of crosslinks with other electrolyte constituents **[11]**.

4.5.1. Carrageenan-Based Electrolytes

Bantang et al. created a sandwich-style DSSC using pure κ-carrageenan as the electrolyte **[57]**. They investigated how the performance of electrolyte gel was affected by the content of κ-carrageenan and several preparation solvents (DMSO and acrylonitrile/water). It was discovered that adding carrageenan up to 1 weight percent of κ-carrageenan increased the conductivity of these κ-carrageenan-based electrolytes because it increased the number of functional groups that were available. On the other hand, conductivity decreased and entanglement increased at a greater concentration chain.

4.5.2. Carrageenan Derivative-Based Electrolytes

It has been found that carboxylation of κ-carrageenan increases the conductivity significantly when compared to pristine carrageenan [20]. This is because it adds more oxygen to the polymer, which makes it easier to establish coordination bonds with more cations. Bella et al. also observed that carboxymethyl κ-carrageenan (CMKC) had conductivity that was two orders of magnitude higher than that of pure carrageenan [58]. For the electrolyte containing 40% NaI and 30% EC, they discovered the maximum conductivity of 3.25 mS/cm. Similar to κ-carrageenan, carboxymethylation can also improve the conductivity and efficiency of hybrid κ/ι-carrageenan and ι-carrageenan-based electrolytes. In addition to providing additional oxygen for ion transport, carboxymethylation decreases the gel's crystalline structure, which improves the polymer's segmental motion. By carboxylation, for instance, Jumaah et al. were able to raise the efficiency of ι-carrageenan from 2.86×10^{-7} to 2.21×10^{-4} S/ cm **[20]**. Later, Torres et al. **[21]** added salt and plasticizer (NH_4I and glycerol) to carboxymethyl κ/ι-hybrid carrageenan to improve its conductivity.

4.6. Other Biopolymers

A non-polysaccharide biopolymer, gelatin, has also been

utilized as a DSSC electrolyte in addition to the polysaccharide mentioned above. It is a good choice for electrolyte synthesis because of the hydroxyl, carboxylic, and amino functional groups on its polymeric backbone. However, when multiwall carbon nanotubes were present, the efficiency of the gelatin-based electrolytes was 1.35%, 1.96%, and 4.02%, respectively, for gold, graphene oxide, and multiwall carbon nanotubes **[59]**. Using a microporous hybrid polymer of poly(acrylic acid)/ gelatin/polyaniline, a 6.94% efficient quasi-solid state DSSC may also be created **[60]**.

5. CONCLUSION

The electrolyte, the active component that controls the charge transfer, is a key factor in the overall efficiency of DSSCs, which are third-generation solar cells. This review discusses the applications of different electrolytes, such as liquid, gel, and solid. Although non-biodegradable polymer electrolyte greatly increases DSSC stability, it has adverse environmental effects. Natural biopolymers are used in place of these non-biodegradable polymer-based electrolytes to address the environmental problem. The weak conductivity of biopolymers can be addressed by altering them through cross-linking, grafting, or the addition of nanoparticles. It is a promising electrolyte material for DSSC applications due to its intrinsic chemical structure and capacity to blend with other natural or synthetic biopolymers. Although biopolymer-based DSSCs are less efficient than other conventional solar cells like silicon, perovskite, and others, research interest in this technology is growing quickly because of its affordability, accessibility, durability, nontoxicity, and environmentally friendly qualities.

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